SF 298 MASTER COPY KEEP THIS COPY FOR REPRODUCTION PURPOSES

REPORT DOCUMENTATION PAGE

Form Approved
OMB NO. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
2/15/99

3. REPORT TYPE AND DATES COVERED
Final Report; 7/1/96-6/30/98

4. TITLE AND SUBTITLE
Protocols for Creating Atomic and Molecular Quantum Computing Elements

5. FUNDING NUMBERS
DAAH04-96-1-0300

6. AUTHOR(S)
Herschel Rabitz

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)
Department of Chemistry
Princeton University
Princeton, NJ 08544

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES)

U.S. Army Research Office
P.O. Box 12211
Research Triangle Park, NC 27709-2211

10. SPONSORING / MONITORING AGENCY REPORT NUMBER
ARO 35778.1-PH-QC

11. SUPPLEMENTARY NOTES
The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

12a. DISTRIBUTION / AVAILABILITY STATEMENT

Approved for public release; distribution unlimited.

12 b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

Under this grant, a constructive procedure was developed for generating a prescribed unitary transform via the optically driven evolution of a multilevel atom. Assuming a clean separation of the coupled levels, the procedure employed the rotating wave approximation together with a decomposition of a unitary matrix into simpler matrix factors with specified structure. Applications to state preparation and observation were also provided.

14. SUBJECT TERMS

unitary evolution, quantum control, quantum computing

15. NUMBER IF PAGES
19

16. PRICE CODE

17. SECURITY CLASSIFICATION OR REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

TABLE OF CONTENTS

1 Introduction

The preparation of an *a priori* prescribed finite-dimensional unitary transformation via the evolution of an externally driven quantum system is a theoretical and technological challenge which plays an important role in several fields including atomic and molecular manipulation [1, 2, 3, 4], quantum computation [5, 6], quantum cryptography [7]. This paper will describe a systematic technique to generate arbitrary special unitary transformations in a quantum system which draws on concepts from control theory. The results described here are applicable to the broader domain of quantum system manipulation.

The principal concept exploited here is that any special unitary matrix can be decomposed into products of transformations with a particular structure. Similar decompositions have recently been suggested as well. Reck and Zeilinger [8] use the decomposition of any $m \times m$ unitary matrix into a product of simpler ones (i.e., tensor products of an arbitrary 2×2 block and a complementary block consisting of the identity matrix of dimension $(m-2)$ obtained in [9] to produce an optical implementation of quantum cryptography schemes. In any factoring approach one first needs to determine the appropriate structure within the factors and then show that any special unitary matrix can be written as a product of such factors. In this regard, Eberly and Law, [10], considered a decomposition for controlling the quantum state of a cavity field. There is a relationship between their factors and those in the present paper, and the number of factors needed are different. Finally, DiVincenzo and collaborators, have demonstrated that any unitary matrix can be written as a product of unitary matrices which are either a tensor product of an arbitrary 2×2 block and I_{M-2} or a special 4×4 block and I_{M-4} [11]. This latter decomposition was motivated by a multiparticle implementation of quantum logic gates. Here we will consider the fundamental case that the non-trivial sub-block is of size 2×2. Unlike the conventional decomposition where the 2×2 blocks are in U(2), the approach presented in this paper respects the selection rules appropriate to the system. Moreover, it can be tailored to take into account practical laboratory constraints.

The specific ingredients of our approach to creating the unitary matrices has the following steps:

- Employ an atom or other simple quantum system whose energy levels are well separated, such that there are no interfering resonances with any pair of optically coupled levels. Assurance of this condition may call for the introduction of suitable static external fields. Spectral separation permits the system to be controlled by sequentially addressing pairs of levels. Furthermore, this circumstance allows one to employ the rotating wave approximation when considering control field designs. Controllability is also possible under conditions more relaxed than this clean spectral separation, but each such case must be individually analyzed for its applicability.
- Decompose the prescribed $N \times N$ unitary matrix into a product of $N \times N$ unitary matrices which are non-trivial only in a sub-block. Here we will consider the simplest case where the sub-block is of size 2×2. This decomposition will respect the selection rules appropriate to the system, and will also be tailored to take into account the rotating wave approximation and any limitations on the matrix elements of the electric dipole operator, which is assumed to be the coupling operator. The decomposition used in this paper is different from the standard one where the 2×2 block can be any element of $U(2)$ (see [9]).
- An explicit characterization of the exponential of an arbitrary real linear combination of the Pauli matrices will be utilized. This formulation will be useful in parametrizing the 2×2 blocks of the previous steps to provide an explicit design for the laboratory control.

The balance of this paper is organized as follows. Section 2 presents a derivation ofthe decomposition needed for the physical system. Theorem 2.1 demonstrates that any matrix in $SU(2)$ can be decomposed as desired. In addition, it is shown how selection rules can be respected during the decomposition process. Remark 2.1 explains how the number of factors may be minimized. Setion 3 discusses how the concepts in the paper may be

applied to state preparation and observation. Finally the Conclusion section 4 addresses the extent to which the ideas in this paper may be generalized to other applications.

2 Generation of Atomic Scale Unitary Transformations

Consider an atom irradiated by a seuqence of electromagnetic pulses with the aim of guiding its evolution initially from the identity matrix to a desired final unitary matrix.

The atom is assumed to have the following properties:

1. The atom has M accessible levels which are well separated for controllability based on frequency discrimination.

2. The matrix elements of the dipole operator (with respect to the basis consisting of the eigenfunctions of the free Hamiltonian) are all real and the diagonal elements are identically zero. This latter assumption can be relaxed in certain cases.

3. The atom-pulse pair is "controllable" in the following sense. There is always a connection between any two levels, i.e., if selection rules preclude accessing level i directly from j, then there is a set of levels (possibly more than one) forming a ladder such that state i can be accessed at least indirectly from state j.

The multi-level atom will be irradiated by a sequence of tailored pulses. Each of the pulses above will address only a prescribed pair of levels. There is flexibility in the pulse structure and each should be chosen to be as simple as possible. The action of the entire sequence of pulses is described below.

The evolution of the unitary generator is described by :

$$i\dot{U}(t,0) = (H_0 - \mu\epsilon)U(t,0) \tag{2.1}$$

where μ is the induced dipole operator and $\epsilon(t)$ is the control field. Defining $\Omega(t,0)$ by

$$U(t,0) = \exp(-iH_0t)\Omega(t,0)$$

leads to the interaction representation

$$i\dot{\Omega}(t,0) = -\epsilon(t)e^{iH_0t}\mu e^{-iH_0t}\Omega(t,0)$$

The control pulse is taken to be $\epsilon(t) = A(t)cos(\omega t + \phi)$. where ϕ is a phase to be chosen and $A(t)$ is assumed to be slowly time varying compared to ω^{-1}, and the frequency ω is resonant with levels i and j to be coupled.

The $M \times M$ matrix $\epsilon(t)e^{iH_0t}\mu e^{-iH_0t}$ will have all allowed couplings given by μ, but, given the conditions stated earlier, we may neglect all terms exccept the (i,j)th and (j,i)th entries which respectively are $\mu_{ij}e^{i\phi}A(t)$ and $\mu_{ij}e^{-i\phi}A(t)$ (recall that all entries of the matrix representation of μ are assumed to be real and the diagonal entries are all zero).

The pulse is applied for a time t_1 with the only restriction that the integral $\int_0^{t_1} A(t)dt$ be insensitive to a slight change in t_1. Applying the rotating wave approximation yields:

$$\Omega(t_1,0) = V_1, \Omega(0,0)) = Id_M$$

where the matrix V_1 is a $M \times M$ matrix that is the identity except for a 2×2 block, which is the exponential of a 2×2 matrix of the form

$$\begin{pmatrix} 0 & i\gamma_1 \\ i\bar{\gamma_1} & 0 \end{pmatrix} \tag{2.2}$$

$\bar{\gamma_1}$ is the complex conjugate of the complex number γ_1 which has the following polar representation:

$$\gamma_1 = \mu_{ij}e^{i\phi}\int_0^{t_1} A(t)dt \tag{2.3}$$

It will be seen later (Equation (2.5)) that the magnitude of the complex number γ_1 enters into our considerations only as the argument of trigonometric functions. Hence $A(t)$ and ϕ are chosen so that γ_1's magnitude can take any desired value within $[0, 2\pi]$ to achieve the necessary action to generate V_1. This point is useful for practical laboratory considerations.

After the application of this first pulse the unitary generator is given by:

$$U(t_1,0) = e^{-iH_0t_1}V_1$$

Now another pulse of length t_2 is applied which is resonant with only some other allowed pair of levels. We then have:

$$U(t,0) = U(t,t_1)U(t_1,0)$$

Once again setting $U(t,t_1) = e^{-iH_0(t-t_1)}\Omega(t,t_1)$ gives:

$$i\dot{\Omega}(t,t_1) = -\epsilon(t)e^{iH_0(t-t_1)}\mu e^{-iH_0(t-t_1)}\Omega(t,t_1)$$

Premultiplying both sides of the last equation by $e^{iH_0t_1}$, using the rotating wave approximation and then integrating up to time $t = t_2$ yields:

$$e^{iH_0t_1}\Omega(t_2,t_1) = V_2e^{iH_0t_1}\Omega(t_1,t_1)$$

Here V_2 is again a $M \times M$ matrix which is the identity except for a 2×2 block which is the exponential of a 2×2 matrix similar in structure to that in (2.2). As $\Omega(t_1,t_1)$ is the identity matrix, the last equation becomes:

$$\Omega(t_2,t_1) = e^{-iH_0t_1}V_2e^{iH_0t_1}$$

Hence, $U(t_2,t_1) = e^{-iH_0(t_2-t_1)}\Omega(t_2,t_1) = e^{-iH_0t_2}V_2e^{iH_0t_1}$, and

$$U(t_2,0) = U(t_2,t_1)U(t_1,0) = e^{-iH_0t_2}V_2V_1$$

If a total of k such coherently locked pulses are applied (starting at $t = 0$ and lasting for a total period of time $T = t_1 + t_2 + \ldots + t_k$) we get:

$$U(T,0) = e^{-iH_0T}V_kV_{k-1}\ldots V_2V_1 \tag{2.4}$$

where the $V_i, i = 1,\ldots,k$ are $M \times M$ matrices with each being the identity except for a 2×2 block which is the exponential of a 2×2 matrix similar in structure to that in (2.2), with γ_1 replaced by suitable complex numbers γ_i. If it is desired that $U(T,0)$ be a prescribed matrix, V, then the question that needs to be addressed is whether V can be decomposed as in the last equation by a suitable choice of pulses. Equivalently, since e^{-iH_0T} is already known, the question is whether any $M \times M$ unitary matrix can be decomposed as the product $\Pi_{i=1}^{k}V_i$ for some choice of k? If the answer is affirmative then

one can consider optimizing the sequence. This optimization would seek to minimize k, while simultaneously considering the pulse shaping capabilities of the apparatus.

It is necessary to show that V can be written as $e^{-iH_0T}\Pi_{k=1}^{L}V(\gamma_k)$. Equivalently we have to show that every unitary matrix $\tilde{V} = e^{iH_0T}V$ can be written as $\Pi_{k=1}^{L}V(\gamma_k)$. It will be shown below that this can always be achieved if $\tilde{V}$ is in $SU(M)$. As any $\tilde{W} \in U(M)$ can be written as $e^{i\Gamma}\tilde{V}$ with Γ a real scalar and $\tilde{V} \in SU(M)$, this shows that the goal of creating V, up to a scalar phase factor which is irrelevant, has been attained.

In the remainder of the paper we will assume that $V \in SU(M)$ and denote by V^H its Hermitian conjugate. Towards the end of demonstrating that V has the desired factorization, we premultiply V^H by a product of matrices with each factor in the product being a matrix which is the identity except for a 2×2 block of the form $V(\gamma_k)$, so that the result is the identity $M \times M$ matrix.

Before proceeding to the general situation we first illustrate, via examples, how selection rules are respected in the process of reducing V^H to the identity by left-multiplication. Assume that the atom has $M = 4$ levels and that the only allowed transitions are $1 \rightarrow 2$ and $1 \rightarrow 3$ and $1 \rightarrow 4$. Any two levels are still accessible from one another, albeit via level 1.

Denoting $V^H = (a_{ij}), i, j = 1, \ldots, 4$, we will first reduce column 4 to $(0,0,0,1)^T$, then column 3 to $(0,0,1,0)^T$, then column 2 to $(0,1,0,0)^T$ and then finally column 1 to $(1,0,0,0)^T$. The reduction will be achieved by premultiplying V^H by suitable matrices which are tensor products of I_2 with a $V(\gamma_k)$. Notice that this order is not the only possible one. All that is important is that column 1 be reduced last. In order to appreciate this point suppose to the contrary that column 1 has been transformed first to $(1,0,0,0)^T$, thereby leaving a matrix of the form

$$\begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & a_1 & b_1 & c_1 \\ 0 & a_2 & b_2 & c_2 \\ 0 & a_3 & b_3 & c_3 \end{pmatrix}$$

with the 3×3 matrix on the bottom right corner an element of $SU(3)$. Once this is achieved we will not be able to reduce the remaining columns (if any) to unit columns because level 1 will no longer have any population in it to effect any further intermediate transitions required between the remaining levels.

Returning now to the procedure suggested above, consider the reduction of column 4 to the corresponding unit column. We may view the fourth column as a particular state of a 4 level system. First transfer all of the amplitude from the a_{24} element to the a_{14} element by using the $1 \rightarrow 2$ transition. This can be effected by premutiplying V^H by a matrix in $SU(4)$ of the form:

$$\begin{pmatrix} a_1 & b_1 & 0 & 0 \\ c_1 & d_1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

The submatrix $\begin{pmatrix} a_1 & b_1 \\ c_1 & d_1 \end{pmatrix}$ is in $SU(2)$ whose elements encode information about a_{24} and a_{14} (see Remark 2.1 below). In the process of this premultiplication, V^H will be transformed into a special unitary matrix with its $(2,4)$ entry equal to 0. Next use the $1 \rightarrow 3$ transition to transfer all population from the $(3,4)$ entry of the new matrix to its $(1,4)$ entry. This may be effected by premultiplying the new matrix by a matrix in $SU(4)$ of the form:

$$\begin{pmatrix} a_2 & 0 & b_2 & 0 \\ 0 & 1 & 0 & 0 \\ c_2 & 0 & d_2 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}$$

The submatrix $\begin{pmatrix} a_2 & b_2 \\ c_2 & d_2 \end{pmatrix}$ in $SU(2)$ encodes information about the $(3,4)$ and $(1,4)$ entries of the new matrix, as before. This premultiplication will result in a $SU(4)$ matrix whose $(2,4)$ and $(3,4)$ entries are both zero. Finally the $1 \rightarrow 4$ transition is used to transform the the fourth column of the last matrix to the column $(0,0,0,1)^T$. Once again

this may be achieved by premutiplying the last matrix by a matrix in $SU(4)$ of the form

$$\begin{pmatrix} a_3 & 0 & 0 & b_3 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ c_3 & 0 & 0 & d_3 \end{pmatrix}$$

where the matrix $\begin{pmatrix} a_3 & b_3 \\ c_3 & d_3 \end{pmatrix}$ in $SU(2)$ is specifically tailored to achieve the desired effect. After this sequence of premultiplications the net effect is a matrix in $SU(4)$ whose last column is $(0,0,0,1)^T$. Unitarity requires that the 4th row be $(0,0,0,1)$. Special unitarity now forces the remaining 3×3 block, $\tilde{V}$, to be in $SU(3)$. We now proceed to reduce the third column of $\tilde{V}$ to $(0,0,1)^T$ by making use of transition $1 \rightarrow 2$ to transform all the amplitude to level 1, and then using the transition $1 \rightarrow 3$ to reduce the third column of the resultant matrix to $(0,0,1)^T$. As before both of these transitions are achieved by premultiplication by matrices in $SU(4)$ whose last column and last row are $(0,0,0,1)^T$ and $(0,0,0,1)$. The balance of these matrices are tensor products of I_1 and the matrix in $SU(2)$ specifically tailored to achieve the desired effects.

Remark 2.1 Let us consider the structure of the matrices in $SU(2)$ which achieve the desired effects above. For example in nulling out the a_{24} element of V^H, we have several choices for the matrix in $SU(2)$. Let r be the norm of the vector $\begin{pmatrix} a_{14} \\ a_{24} \end{pmatrix}$, and then seek a matrix, $V_{12} \in SU(2)$ such that

$$V_{12} \begin{pmatrix} a_{14} \\ a_{24} \end{pmatrix} = \begin{pmatrix} re^{i\theta} \\ 0 \end{pmatrix}$$

Usually θ can be taken as arbitrary. Only in the last step in converting any column to the corresponding unit vector does θ have to be zero. Fixing θ leads to a unique V_{12} which will achieve the above result. This is just the statement that $SU(2)$ acts transitively on spheres in C^2 of any given radius. However, the freedom in the choice of θ (except in the last step) is useful in minimizing the number of $V(\gamma_k)$'s needed. Indeed it will be shown

below that any matrix in $SU(2)$ is a product of at most 3 matrices of the form $V(\gamma_k)$. So the freedom in choosing θ is useful in minimizing the number of $V(\gamma_k)$'s. In fact, there is a matrix of the form $V(\gamma)$ such that $V(\gamma)\begin{pmatrix} p \\ q \end{pmatrix} = \begin{pmatrix} \sqrt{|p|^2 + |q|^2}e^{i\mu} \\ 0 \end{pmatrix}$ where the complex numbers p and q have polar representations $p = |p| e^{i\mu}$ and $q = |q| e^{i\zeta}$. Notice that the phase of p is the same as that of $V(\gamma)\begin{pmatrix} p \\ q \end{pmatrix}$. This will be demonstrated in Lemma (2.1) later. *Thus not insisting that θ be zero (except in one step) reduces the number of $V(\gamma_k)$'s to be used and thus the number of pulses to be applied.*

The general case of an M-level atom can be handled by directly extending the above procedure to express any matrix in $SU(M)$ as a product of matrices which are tensor products of I_{M-2} and an element of $SU(2)$. Thus all that remains is to show that every element of $SU(2)$ can be written as a product of $V(\gamma_k)$'s and this is achieved by the theorem below.

Theorem 2.1 *Any matrix $V \in SU(2)$ can be expressed as the product $\Pi_{k=1}^{l} V(\gamma_k)$. Furthermore, l can be taken to be no more than three.*

Proof: The proof proceeds via the following steps:

1. Suppose $\gamma = re^{i\phi}$ is given. Then

$$W(\gamma) = \exp\begin{pmatrix} 0 & i\gamma \\ i\bar{\gamma} & 0 \end{pmatrix}$$

Let $R^2 = \alpha^2 + \beta^2$ (thus $R = 2r$). Employing the formula for the exponential of an arbitrary real linear combination of the Pauli matrices given in the Appendix, with $\delta = 0$, $\gamma = \frac{\alpha - i\beta}{2}$ we get:

$$V(\gamma) = \begin{pmatrix} \cos\frac{R}{2} & 0 \\ 0 & \cos\frac{R}{2} \end{pmatrix} + \frac{i\sin\frac{R}{2}}{\frac{R}{2}}\begin{pmatrix} 0 & \frac{\alpha-i\beta}{2} \\ \frac{\alpha+i\beta}{2} & 0 \end{pmatrix}$$

$$= \begin{pmatrix} \cos\frac{R}{2} & 0 \\ 0 & \cos\frac{R}{2} \end{pmatrix} + \frac{i\sin r}{r}\begin{pmatrix} 0 & re^{i\phi} \\ re^{-i\phi} & 0 \end{pmatrix}$$

Hence,

$$V(\gamma) = \begin{pmatrix} \cos\, r & ie^{i\phi}\sin\, r \\ ie^{-i\phi}\sin\, r & \cos\, r \end{pmatrix} \tag{2.5}$$

2. For $S \in SU(2)$, the following holds:

$$S = \begin{pmatrix} e^{i\lambda}\cos\alpha & e^{i\mu}\sin\alpha \\ e^{i(\pi-\mu)}\sin\alpha & e^{-i\lambda}\cos\alpha \end{pmatrix}$$

with $\lambda > 0$ and $\mu > \frac{\pi}{2}$, and some real α.

Proof: If $V = \begin{pmatrix} a & b \\ c & d \end{pmatrix}$, then the rows of of V are orthogonal and $detV = 1$. So, if $a = \cos\alpha e^{i\lambda}$ and $b = \sin\alpha e^{i\mu}$, then V admits the above description. Here λ and μ are chosen to satisfy the inequality constraints required. Clearly λ can always be chosen positive. If μ does not already satisfy the required inequality constraints, then we may write $b = e^{i(\pi+\mu)}\sin(-\alpha)$, and then take $\pi + \mu$ as the new μ, and $-\alpha$ as the new α. Since the original μ could always be chosen positive, this achieves the desired inequality constraint on μ. Also, as $\cos(-\alpha) = \cos(\alpha)$, a is unaffected by this process.

3. The number of $W(\gamma)$'s required for writing S as a product of them can be taken to be no more than three.

Proof: Let $\gamma_1 = \alpha e^{i\theta}$, where $\theta = \lambda + \mu - \frac{\pi}{2}$. Then $S = W(\gamma_1)\begin{pmatrix} e^{i\lambda} & 0 \\ 0 & e^{-i\lambda} \end{pmatrix}$.

Now $\begin{pmatrix} e^{i\lambda} & 0 \\ 0 & e^{-i\lambda} \end{pmatrix} = W(\gamma_2)W(\gamma_3)$ where $\gamma_2 = \frac{\pi}{2}e^{i\sigma_1}$ and $\gamma_3 = \frac{\pi}{2}e^{i\sigma_2}$ where σ_1 and σ_2 are any real numbers such that $\lambda = \sigma_1 - \sigma_2 + \pi$. This completes the proof of Theorem 2.1.

Finally, the lemma which was used in Remark (2.1) follows:

Lemma 2.1 *Given a vector* $\begin{pmatrix} p \\ q \end{pmatrix}$ *in* C^2 *there exists a complex number* γ *such that:*

$$W(\gamma)\begin{pmatrix} p \\ q \end{pmatrix} = \begin{pmatrix} \sqrt{|p|^2 + |q|^2} e^{i\mu} \\ 0 \end{pmatrix}$$

Proof: Choose $\gamma = \alpha e^{i\theta}$, where $\cos\alpha = \frac{|p|}{|p|^2+|q|^2}$ and $\theta = \mu - \lambda - \frac{\pi}{2}$.

Clearly

$$\sin\alpha = \frac{|q|}{|p|^2 + |q|^2}$$

Thus,

$$W(\gamma)\begin{pmatrix} p \\ q \end{pmatrix}$$

$$= \begin{pmatrix} \cos\alpha & i\sin\alpha e^{i(\mu-\lambda-\frac{\pi}{2})} \\ i\sin\alpha e^{i(\lambda-\mu+\frac{\pi}{2})} & \cos\alpha \end{pmatrix} \begin{pmatrix} |p| e^{i\mu} \\ |q| e^{i\lambda} \end{pmatrix}$$

$$= \begin{pmatrix} |p|^2 e^{i\mu} + i |q|^2 e^{i(\mu-\frac{\pi}{2})} \\ 0 \end{pmatrix}$$

$$= \begin{pmatrix} \sqrt{|p|^2 + |q|^2} e^{i\mu} \\ 0 \end{pmatrix}$$

3 Applications to State Preparation and Observation

This section will consider applications of the generation of special unitary matrices to a) state preparation, and b) state observation. Case a) will be discussed from the perspective of controllability, [12] and case b) aims to show that the preparation of certain sequences of special unitary matrices leads to the determination of the state of the system.

- State Preparation: The case of state preparation is implicitly present in a previous paper [12], where the problem of controllability of molecular systems was analyzed by considering the system defined by the corresponding unitary generator. However, a slight refinement is needed to utilize that analysis, since in this paper we have only shown how special unitary matrices may be prepared. Given two vectors u and v on the unit sphere of C^M there is an entire family of unitary matrices U such that $Uu = v$. To construct them we first consider the unitary matrices U_u and U_v, where U_u and U_v have as their first columns the vectors u and v. The remaining columns can be any vectors which render U_u and U_v unitary (e.g., those obtained by applying the Gram-Schmidt process). Clearly $U_u p = u$ and $U_v p = v$ where $p = (1, 0, \ldots, 0)^T$. Then the matrix $U = U_v U_u^H$ satisfies $Uu = v$.

  Now consider the case where the matrix acting on u to give v is desired to be special unitary. Construct $S_u = AU_u^H$ and $S_v = U_v B$ according to $A = \mathrm{diag}(1, e^{i\delta_u}, 1, \ldots, 1)$, where $e^{i\delta_u} = \mathrm{det} U_u$, and $B = \mathrm{diag}(1, e^{-i\delta_v}, 1, \ldots, 1)$ with $e^{i\delta_v} = \mathrm{det} U_v$. Then both S_u and S_v are special unitary and thus the product $S = S_v S_u$ is also special unitary and satisfies $Su = v$, since $Uu = v$. Hence by preparing the special unitary matrix S and acting on the initial state u it is possible to create the desired state v. In most situations $u = (1, 0, \ldots, 0)^T$.

  It would be interesting to examine this approach for initial state creation for low dimensional systems. The ability to create initial states for low dimensional systems can be combined with other approaches such as optimal control [2, 13, 3, 4] to create desired states for higher dimensional systems.

- State Observation: State observation is fundamental to control engineering [14], and it is equally significant for quantum systems [15]. The ability to determine that a desired state has been actually obtained is needed to assess the success of the control process and its use in a variety of applications. We assume that the measurement process can determine the population of some particluar level N. Thus,

if $\psi_{\text{Final}} = \sum_{i=1}^{M} c_i e^{i\phi_i}\psi_i$, where $\psi_i, i = 1, \ldots, M$ is some basis (e.g., eigenfunctions of the free Hamiltonian), then we are able to measure $\mid c_N \mid^2$. By preparing the special unitary matrix which results in the exchange of populations between levels N and n, it is possible to measure $\mid c_n \mid^2$, for all $n = 1, \ldots, M$. The only remaining quantities to be determined are the phases $\phi_i, i = 1, \ldots, M$. Since only relative phases are relevant, we assume that the reference phase ϕ_N is 0. Next the special unitary matrix which is the identity except for the following 2×2 block in the (N, n) position is prepared:

$$\frac{1}{\sqrt{2}}\begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}$$

When applied to ψ_{Final} it results in a vector which is identical to ψ_{Final} except that c_N and c_n are replaced by $\zeta_N = \frac{c_N + c_n}{\sqrt{2}}$ and $\zeta_n = \frac{c_N - c_n}{\sqrt{2}}$. By assumption, we can readout $\mid \zeta_N \mid^2$. Since,

$$\mid \zeta_N \mid^2 = \frac{1}{2}\{\mid c_N \mid^2 + \mid c_n \mid^2 + c_N c_n^* + c_N^* c_n\}$$

it is possible to determine $M_n = \frac{|\zeta_N|^2 - \frac{1}{2}(|c_N|^2 + |c_n|^2)}{2|c_N|\,|c_n|} = \frac{e^{i(\phi_N - \phi_n)} + e^{-i(\phi_N - \phi_n)}}{2} = \cos(\phi_N - \phi_n)$. However, this does not determine ϕ_n since the cosine function is double valued on $(0, 2\pi)$.

To remedy this latter problem we prepare the special unitary matrix which is the tensor product of I_{M-2} and the following 2×2 block in the (N, n) position:

$$\frac{1}{\sqrt{2}}\begin{pmatrix} 1 & i \\ i & 1 \end{pmatrix}$$

When applied to ψ_{Final}, the transformation results in a vector which is identical to ψ_{Final} except that c_N and c_n are replaced by $\zeta_N' = \frac{c_N + ic_n}{\sqrt{2}}$ and $\zeta_n' = \frac{ic_N + c_n}{\sqrt{2}}$. Once again, by assumption we are able to measure $\mid \zeta_N' \mid^2$. Since the quantity

$$M_n' = \frac{-ie^{i(\phi_N - \phi_n)} + ie^{-i(\phi_N - \phi_n)}}{2} = \sin(\phi_N - \phi_n)$$

can be measured, it is clear that $(\phi_N - \phi_n)$ can be determined for each n, and hence so can $\phi_n, n = 1, \ldots, M$.

A total of $3(M-1)$ measurements are required on a sequence of controlled systems for the $2(M-1)$ unknowns $|c_i|^2, \phi_i$. The above scheme is a special case of determining the elements of a density matrix via projection measurements developed in [15].

4 Conclusions

In this paper a constructive scheme for the creation of any special unitary matrix has been analyzed. Although the physical system used here is a single particle interacting with an electromagnetic field, there are some general features to the scheme which are relevant for other quantum systems [16]. These features are:

- The quantum system in question can be controlled by addressing subsystems of dimension two sequentially.
- The logarithm of the unitary generator, obtained after the controlling mechanism is applied to any of these two dimensional subsystems, lies in the span of any two of the Pauli matrices.

The first point above suggests that we represent our desired special unitary matrix as the products of factors which are non-trivial only in a 2×2 block. If no special structure on this 2×2 block is desired, then this can always be achieved, [9]. If a special structure is desired, then a preliminary question is what special structures are feasible? The second point yields the answer. The reason is that the Lie algebra generated by any two of the Pauli matrices equals the entire Lie algebra of $SU(2)$. Thus, if the second point is valid for the quantum system being studied then any matrix in $SU(2)$ can be decomposed, in principle at least, into a product of matrices with the prescribed special structure. The very important practical questions of obtaining this decomposition in an algorithmic fashion and also minimizing the number of factors in the decomposition depend on the particular system. However, if the quantum system is such that in the second point above the logarithm belongs to the span of $i\sigma_x$ and $i\sigma_y$, then the constructive algorithm introduced here is applicable to it as well.

Although quantum computation, [5, 6], was not the primary focus of this work, its needs provide useful background. Any proposal to fabricate a quantum computer encompasses three steps: i) Preparation of the initial state which will serve as the input; ii) Preparation of the unitary transformation which will function as the logic gate; and iii) Reading the final output. With every concrete protocol proposed for a quantum computer the physical means for achieving these 3 steps must be specified. In addition the nature of each step depends on the particular problem or quantum algorithm. This paper considered a single atom (i.e., a particle in the limit of one electron) as a realization. There are many practical limitations associated with this choice (as is the case with any other choice thus far presented), but there are also strong arguments to suggest that an atom would be an ideal system for practical laboratory study. Single particles are attractive from the point of view of isolating the system from external influences, and thereby enhancing the lifetime (provided states of sufficient radiative lifetime are involved). Furthermore, both the theoretical and experimental aspects of controlling molecular and atomic dynamics are advancing very rapidly. All three steps above can be subsumed into the problem of creating a prescribed unitary matrix from the evolution of a controlled atom. It is possible to achieve entanglement within a single molecule by simultaneously manipulating two degrees of freedom such as rotation and vibration, or radial and angular degrees of freedom in a single atom etc., A further critical matter concerns the structure of the algorithms used in quantum computation. The current algorithms are designed to work with multiple particles. However, this circumstance does not preclude working with multilevel systems The nature of the state observation is also very important. Section 3 considered full state observation, but other less demanding observations are desirable in keeping with the quantum computation algorithms proposed so far. These topics go beyond the scope of this paper, but the work here should provide motivation for exploring these questions further.

Finally, it would be interesting to examine the field of molecular control, [2, 13]¡ from the perspective of this paper. Most problems in molecular control can be cast as state

preparation for mutilevel systems. Section 3 showed that this goal can be subsumed into the problem of creating one out of a family of special unitary matrices. The main feature of our algorithm is that it does not require any costly computations, although the pulse sequenxce still needs to be determined.

5 Appendix: Exponential of Pauli Matrices

Since no reference was found for the result below, a proof is being included for the sake of completeness. It facilitates obtaining decompositions of unitary matrices adapted to various constraints in the control pulse.

Theorem 5.1 *Let* $\sigma_x = \frac{1}{2}\begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$, $\sigma_y = \frac{1}{2}\begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$ *and* $\sigma_z = \frac{1}{2}\begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$ *be the Pauli matrices. Then we have*

$$expi(\alpha(t)\sigma_x+\beta(t)\sigma_y+\delta(t)\sigma_z) = \cos(\frac{\sqrt{\lambda(t)}}{2})I_2+\frac{2i}{\sqrt{\lambda(t)}}\sin(\frac{\sqrt{\lambda(t)}}{2})(\alpha(t)\sigma_x+\beta(t)\sigma_y++\delta(t)\sigma_z)$$

where $\alpha(t), \beta(t)$ *and* $\delta(t)$ *are real valued functions of time, and*

$$\lambda(t) = (\alpha(t)^2 + \beta(t)^2 + \delta(t)^2)$$

Proof: First, a direct calculation shows that $\sigma_x^2 = \sigma_y^2 = \sigma_z^2 = \frac{1}{4}I_2$; $\sigma_x\sigma_y = \frac{i}{2}\sigma_z = -\sigma_y\sigma_x$; $\sigma_y\sigma_z = \frac{i}{2}\sigma_x = -\sigma_z\sigma_y$; and that $\sigma_z\sigma_x = \frac{i}{2}\sigma_y = -\sigma_x\sigma_z$.

Hence,

$$(i\alpha\sigma_x + i\beta\sigma_y + i\delta\sigma_z)^2 = (i\alpha\sigma_x + i\beta\sigma_y + i\delta\sigma_z)(i\alpha\sigma_x + i\beta\sigma_y + i\delta\sigma_z)$$

$$= -[\alpha^2\sigma_x^2 + \alpha\beta\sigma_x\sigma_y + \alpha\delta\sigma_x\sigma_z + \alpha\beta\sigma_y\sigma_x + \beta^2\sigma_y^2 + \beta\delta\sigma_y\sigma_z + \alpha\delta\sigma_z\sigma_x + \delta\beta\sigma_z\sigma_y + \delta^2\sigma_z^2]$$

$$= \sum_{n=0}^{\infty} \frac{1}{(2n)!}(\frac{-\lambda}{4}I_2)^n + \sum_{n=0}^{\infty} \frac{1}{(2n+1)!}(\frac{-\lambda}{4}I_2)^n(i\alpha\sigma_x + i\beta\sigma_y + i\delta\sigma_z)$$

$$= \frac{-1}{4}(\alpha^2 + \beta^2 + \gamma^2)I_2$$

Hence

$$e^{i(\alpha I_x+\beta I_y+\gamma I_z)} = \sum_{n=0}^{\infty} \frac{1}{(2n)!}(i\alpha I_x + i\beta I_y + i\gamma I_z)^{2n} + \sum_{n=0}^{\infty} \frac{1}{(2n+1)!}(i\alpha I_x + i\beta I_y + i\gamma I_z)^{2n+1}$$

$$==\sum_{n=0}^{\infty}\frac{1}{(2n)!}(\frac{-\lambda}{4}I_2)^n+\sum_{n=0}^{\infty}\frac{1}{(2n+1)!}(\frac{-\lambda}{4}I_2)^n(i\alpha I_x+i\beta I_y+i\gamma I_z)$$

$$=\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n)!}(\frac{\sqrt{\lambda}}{2})^{2n}I_2+i\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n+1)!}(\frac{\sqrt{\lambda}}{2})^{2n}(\alpha I_x+\beta I_y+\gamma I_z)$$

$$=\cos(\sqrt{\lambda})I_2+\frac{2i}{\sqrt{\lambda}}(\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n+1)!}(\frac{\sqrt{\lambda}}{2})^{2n+1}(\alpha I_x+\beta I_y+\gamma I_z)$$

$$=\cos(\sqrt{\lambda})I_2+\frac{2i}{\sqrt{\lambda}}\sin(\frac{\sqrt{\lambda}}{2})(\alpha I_x+\beta I_y+\gamma I_z).$$

which is the desired result.

References

[1] R. Gordon and S. Rice, *Ann. Rev. Phys. Chem.*, **48**, 601 (1997).

[2] W. Warren, H. Rabitz and M. Dahleh, Science **259**, 1581 (1993).

[3] K. Wilson et. al., *J. Phys. Chem.*, **97**, 2320 (1995).

[4] A. Butkovsky and Y. Samoilenko, *Control of Quantum Mechanical Processes and Systems*, Kluwer Academic, Dordrecht, 1990.

[5] R. Feynman, Optics News, **11**, 11 (1985)

[6] P. Shor in Proceedings of the 25th Annual ACM Symposium on Foundations of Computer Science, S. Goldwasser, ed., IEEE Computer Society, Los Alamitos, CA, 1994, Pg 124.

[7] A. Sudberry, Nature, **362**, 586 (1993).

[8] M. Reck, A. Zeilinger, H. Bernstein and P. Bertani, *Phys Rev Lett*, **73**, 58 (1994).

[9] F. Murnaghan, *The Unitary and Rotation Groups*, Spartan Books, 1960.

[10] C. Law and J. Eberly, *Phys. Rev. Lett.*, **76**, 1055, (1996).

[11] P. DiVincenzo,*Phys. Rev A.*, **51**, 1015 (1995).

[12] V. Ramakrishna, M. Salapaka, M. Dahleh, H. Rabitz and A. Peirce, *Phys. Rev A.*, **51**, 960 (1995).

[13] S. Shi and H. Rabitz, *J. Chem. Phys.*, **92**, 364 (1990).

[14] J. Zabcyk, *Mathematical Control Theory*, Birkhauser, Basel, 1990.

[15] O. Steuernagel and J. Vaccaro, *Phys. Rev. Lett.*, **75**, 3201(1995).

[16] V. Ramakrishna, R. Ober and H. Rabitz, "Explicit Generation of States in Quantum Control", submitted to the Proceedings of the Mathematical Theory of Netwroks and Systems Conference, July 6th -10th, 1998, Padova, Italy.

OTHER REQUESTED INFORMATION

Publications Supported Under This Grant.

- Explicit Generation of Unitary Transformations in a Single Atom, V. Ramakrishna, R. Ober, X. Sun, O. Steuernagel, H. Rabitz, and J. Botina, *Phys. Rev. A*, submitted.

Scientific Personnel Supported by this Project.

H. Rabitz; V. Ramakrishna, Assistant Professor, University of Texas; and W. Zhu, a post-doctoral associate.

Report of Inventions. None